Mechanical characterization of brain tissue in compression at dynamic strain rates

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Large acoustoelastic effect

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Abstract

Classical acoustoelasticity couples small-amplitude elastic wave propagation to an infinitesimal pre-deformation, in order to reveal and evaluate non-destructively third-order elasticity constants. Here, we see that acoustoelasticity can be also be used to determine fourth-order constants, simply by coupling a small-amplitude wave with a small-but-finite pre-deformation. We present results for compressible weakly nonlinear elasticity, we make a link with the historical results of Bridgman on the physics of high pressures, and we show how to determine “D”, the so-called fourth-order elasticity constant of soft (incompressible, isotropic) solids by using infinitesimal waves.

Keywords: acousto-elasticity, high pressures, large pre-tension, elastic constants
1 Introduction

Acoustoelasticity is now a well established experimental technique used for the non-destructive measurement of third-order elasticity (TOE) constants of solids, and its principles can be found in standard handbooks of physical acoustics, such as Pao et al. (1984); Kim and Sachse (2001). The underlying theory, however, is quite intricate, and over the years several authors have produced different and irreconcilable expressions for the shift experienced by the wave speed when elastic wave propagation is coupled to a pre-strain. A common mistake found in the literature consists of the implicit assumption that since a small pre-strain and a small-amplitude wave are described by linearized equations, they can be superposed linearly. With that point of view, the coupling between the two phenomena is of higher order and can be neglected in the first approximation. The flaw in that reasoning is simply that these phenomena are successive, not linearly superposed. Experiencing an infinitesimal pre-strain from a stress-free configuration is a linear process; propagating an infinitesimal wave in a stress-free configuration is another linear process. But in acoustoelasticity, the wave travels in a pre-stressed, not a stress-free solid, and the laws of linear elastodynamics must be adapted to reflect this fact. The final outcome of this analysis is that not only second-order, but also TOE constants appear in the expression for the speed of an acoustoelastic wave.

For example, a longitudinal wave travelling in a solid subject to a hydrostatic stress $\sigma I$ will propagate with the speed $v_{0L}$ given by

$$\rho v_{0L}^2 = \lambda + 2\mu - \frac{7\lambda + 10\mu + 2A + 10B + 6C}{3\lambda + 2\mu} \sigma,$$  \hfill (1.1)

where $\rho$ is the mass density in the unstressed configuration, $\lambda$ and $\mu$ are the (second-order) Lamé coefficients, and $A, B, C$ are the (third-order) Landau coefficients (Landau and Lifshitz, 1986). This equation was correctly established as early as 1925 by Brillouin and confirmed in 1953 by Hughes and Kelly, although several erroneous expressions have appeared in between and since (see, e.g., Birch (1938); Tang (1967)).

From our contemporary perspective, the easiest way to re-establish and further this expression is to rely on the modern theory of incremental (also known as small-on-large) elasticity, which presents compact expressions for the tensor of instantaneous elastic moduli, $A_0$, given by its components

$$\begin{align*}
A_{0iijj} &= J^{-1} \lambda_i \lambda_j W_{ij}, \\
A_{0ijij} &= J^{-1} \frac{\lambda_i W_i - \lambda_j W_j}{\lambda_i^2 - \lambda_j^2} \lambda_i^2, \\
A_{0ijji} &= J^{-1} \frac{\lambda_j W_j - \lambda_i W_i}{\lambda_j^2 - \lambda_i^2} \lambda_i \lambda_j, \\
A_{0ijij} &= J^{-1} (\lambda_i^2 W_i - \lambda_i \lambda_j W_{ij} + \lambda_i W_i) / 2, \\
A_{0ijji} &= J^{-1} (\lambda_j^2 W_i - \lambda_i \lambda_j W_{ij} - \lambda_j W_i) / 2,
\end{align*}$$  \hfill (1.2)

in the coordinate system aligned with the principal axes of pre-strain, which, since the material is isotropic, coincide with the principal axes of pre-stress. Here, $W$ is the strain-energy density per unit volume, which is a symmetric function of the principal stretches.
\( \lambda_1, \lambda_2, \lambda_3 \) of the deformation, \( J = \lambda_1\lambda_2\lambda_3, \) \( W_i = \partial W / \partial \lambda_i \) and \( W_{ij} = \partial^2 W / \partial \lambda_i \partial \lambda_j, \ i, j \in \{1, 2, 3\}; \) see, for example, Ogden (1997, 2007) for details. The corresponding principal Cauchy stresses are given by \( \sigma_i = J^{-1} \lambda_i W_i, \ i = 1, 2, 3. \)

It is worth noting here for later reference that it follows from (1.2) that

\[
A_{0ijij} - A_{0jiji} = \sigma_i - \sigma_j, \quad i \neq j. \tag{1.3}
\]

A small-amplitude body wave may travel at speed \( v \) in the direction of the unit vector \( \mathbf{n} \) with polarization in the direction of a unit vector \( \mathbf{m} \) provided the eigenvalue problem

\[
Q(n) \mathbf{m} = \rho v^2 \mathbf{m} \tag{1.4}
\]

is solved with \( \rho v^2 > 0 \), where \( Q(n) \) is the acoustic tensor, which has components

\[
Q_{ij}(n) = J A_{0piqj} n_p n_q. \tag{1.5}
\]

Note that sometimes the acoustic tensor is defined by (1.5) without the factor \( J \), in which case the \( \rho \) in (1.4) would be replaced by the density in the deformed configuration, i.e. \( J^{-1} \rho \).

Notice that in this theory the amplitude of the acoustic wave is infinitesimal, but that there is no restriction on the choice of \( W \) or on the magnitude of the underlying pre-strain because we are in the context of finite nonlinear elasticity. It follows that the expressions can be specialized in various ways, in particular to weakly nonlinear elasticity (where \( W \) is expanded in terms of some measure of strain) and to special pre-strains.

Hence, to access the TOE constants, we take \( W \) as

\[
W = W_{TOE} \equiv \frac{\lambda}{2} I_1^2 + \mu I_2 + \frac{A}{3} I_3 + B I_1 I_2 + \frac{C}{3} I_1^3, \tag{1.6}
\]

where \( I_n = \text{tr}(E^n), \ n = 1, 2, 3, \) are invariants of the Green–Lagrange strain tensor \( E \). The components (1.2) can then be expanded to the first order in terms of, for example, the volume change \( \varepsilon \equiv J - 1 \) in the case of a hydrostatic pressure, or of the elongation \( \varepsilon_1 \equiv \lambda_1 - 1 \) in the case of uniaxial tension.

To access fourth-order elasticity (FOE) constants, we take \( W \) as

\[
W = W_{FOE} \equiv W_{TOE} + E I_1 I_3 + F I_1^2 I_2 + G I_2^2 + H I_1^4, \tag{1.7}
\]

where \( E, F, G, H \) are the FOE constants and we push the expansions of the wave speed up to the next order in the strain. This is what we refer to as the large acoustoelastic effect. In fact, the main purpose of this investigation is to provide a theoretical backdrop to the current drive to determine experimentally the FOE constants of soft solids such as isotropic tissues and gels in order to improve acoustic imaging resolution (see, e.g., Hamilton et al. (2004); Zabolotskaya et al. (2004, 2007); Gennisson et al. (2007); Rénier et al. (2007); Jacob et al. (2007); Rénier et al. (2008b,a); Mironov et al. (2009)).

Because soft solids are often treated as incompressible, so that the constraint \( J \equiv 1 \) must be satisfied, we shall also consider the expressions for the TOE and FOE strain-energy densities in their incompressible specializations, specifically

\[
W = \mu I_2 + \frac{A}{3} I_3 \tag{1.8}
\]
for TOE incompressibility and
\[ W = \mu I_2 + \frac{A}{3} I_3 + D I_2^2 \]  
(1.9)

for FOE incompressibility. Hence, in the transition from compressible to incompressible elasticity, the number of second-order constants goes from two to one, of third-order constants from three to one, and of fourth-order constants from four to one. This was established by Hamilton et al. in 2004, although it can be traced back to Ogden in 1974 and even earlier, to Bland in 1969. Specifically, in that transition we note that the elasticity constants behave as (Destrade and Ogden, 2010)
\[ \lambda \rightarrow \infty, \quad \mu \rightarrow E_Y/3, \]  
(1.10)

for the second-order constants, where \( E_Y \) is the (finite) Young’s modulus,
\[ A/\mu = \mathcal{O}(1), \quad B/\mu = \mathcal{O}(\lambda/\mu), \quad C/\mu = \mathcal{O}(\lambda^2/\mu^2) \]  
(1.11)

for the third-order constants,
\[ E/\mu = \mathcal{O}(\lambda/\mu), \quad F/\mu = \mathcal{O}(\lambda^2/\mu^2), \quad G/\mu = \mathcal{O}(\lambda/\mu), \quad H/\mu = \mathcal{O}(\lambda^3/\mu^3) \]  
(1.12)

for the fourth-order constants, and \((C + F)/\mu = \mathcal{O}(\lambda/\mu)\) for a combination of second- and third-order constants. Some constants have the explicit limiting behaviour
\[ B/\lambda \rightarrow -1, \quad E/\lambda \rightarrow 4/3, \quad G/\lambda \rightarrow 1/2, \quad B + G + \lambda/2 \rightarrow D, \]  
(1.13)

where \( D \) remains a finite quantity, of the same order of magnitude as \( \mu \); see Destrade and Ogden (2010) for analysis of the behaviour of \( A, B, \ldots, H \) in the incompressible limit and the connection with \( D \). Note that the limiting behaviour for \( E \) in terms of the initial Poisson’s ratio \( \nu \) and Young’s modulus \( E_Y \) is thus \((1 - 2\nu)E \rightarrow 4E_Y/9\). This was shown in Destrade and Ogden (2010), but mistyped in equations (49) and (89) therein as \((1 - 2\nu)E \rightarrow 4E_Y/3\).

In this paper, we treat in turn the case of hydrostatic pre-stress (Section 2) and of uniaxial pre-stress (Section 3), and we provide expansions of the body wave speeds up to the second order in the pre-strain, and also in the pre-stress, for compressible solids and in the relevant incompressible limits.

2 Hydrostatic pressure

Consider first a cuboidal sample of a compressible solid with sides of lengths \((L_1, L_2, L_3)\) in its (unstressed) reference configuration. We define the reference geometry in terms of Cartesian coordinates \((X_1, X_2, X_3)\) by \(0 \leq X_i \leq L_i, i = 1, 2, 3\). The material is then subject to a pure homogeneous strain \(x_1 = \lambda_1 X_1, x_2 = \lambda_2 X_2, x_3 = \lambda_3 X_3\) and deformed into the cuboid \(0 \leq x_i \leq l_i, i = 1, 2, 3\), where \((x_1, x_2, x_3)\) are the Cartesian coordinates in the deformed configuration, and the constants \(\lambda_1, \lambda_2, \lambda_3\) are the principal stretches of the deformation. We now specialize the deformation to a pure dilatation so that \(\lambda_1 = \lambda_2 = \lambda_3 = J^{1/3}\). Since the material is isotropic the Cauchy stress \(\mathbf{\sigma}\) is spherical, \(\mathbf{\sigma} = \sigma \mathbf{I}\) say, with \(\sigma > 0 \) \((< 0)\) corresponding to hydrostatic tension (pressure).
The pre-stress is computed as $\sigma = J^{-1} \lambda_1 W_1$ or, more conveniently, as $\sigma = \tilde{W}'(J)$, where $\tilde{W}(J) \equiv W(J^{1/3}, J^{1/3}, J^{1/3})$. Up to the second order in the volume change $\varepsilon \equiv J^{-1}$, we find

$$\sigma = \kappa \varepsilon + \frac{1}{2} \tilde{W}''(1) \varepsilon^2,$$

(2.1)

where $\kappa = \tilde{W}'(1) = \lambda + 2\mu / 3$ is the infinitesimal bulk modulus, and

$$\tilde{W}''(1) = -\kappa + \frac{2}{9} (A + 9B + 9C).$$

(2.2)

(It is a simple matter to check that the expansion (2.1) is equivalent to one first established by Murnaghan (1951).) Conversely, the volume change is expressed in terms of the hydrostatic stress as

$$\varepsilon = \frac{1}{\kappa} \sigma - \frac{\tilde{W}''(1)}{2\kappa^3} \sigma^2.$$

(2.3)

Here the pre-stress does not generate preferred directions and the solid remains isotropic. It follows that two waves may propagate in any direction, one longitudinal, with speed $v_L$, and one transverse, with speed $v_T$, given by

$$\rho v_L^2 = \lambda + 2\mu + a_L \varepsilon + b_L \varepsilon^2,$$

$$\rho v_T^2 = \mu + a_T \varepsilon + b_T \varepsilon^2,$$

(2.5)

where

$$a_L = \frac{7}{3} \lambda + \frac{10}{3} \mu + \frac{2}{3} A + \frac{10}{3} B + 2C, \quad a_T = \lambda + 2\mu + \frac{1}{3} A + B,$$

(2.6)

are the coefficients for the classical (linear) acoustoelastic effect, and

$$b_L = \frac{13}{18} \lambda + \frac{7}{9} \mu + \frac{8}{9} A + \frac{44}{9} B + \frac{10}{3} C + \frac{8}{3} E + \frac{16}{3} F + \frac{20}{9} G + 12H,$$

$$b_T = \frac{1}{2} \lambda + \frac{5}{9} \mu + \frac{1}{2} A + \frac{13}{6} B + C + E + F + \frac{2}{3} G$$

(2.7)

are the coefficients of the large (quadratic) acoustoelastic effect.

Alternatively, we may use (2.3) to express the wave speeds in terms of the pre-stress rather than the pre-strain, as

$$\rho v_L^2 = \lambda + 2\mu + c_L \sigma + d_L \sigma^2,$$

$$\rho v_T^2 = \mu + c_T \sigma + d_T \sigma^2,$$

(2.8)

thus recovering the classical formulas

$$c_L = \frac{a_L}{\kappa}, \quad c_T = \frac{a_T}{\kappa},$$

(2.9)

for the (linear) acoustoelastic effect (Hughes and Kelly, 1953), and establishing the formulas

$$d_L = \frac{b_L}{\kappa^2} - \frac{a_L \tilde{W}''(1)}{2\kappa^3}, \quad d_T = \frac{b_T}{\kappa^2} - \frac{a_T \tilde{W}''(1)}{2\kappa^3}$$

(2.10)
for the large (quadratic) acoustoelastic effect, where $\hat{W}'''(1)$ is given in (2.2).

For an application of these results, we turn to the classical data of Hughes and Kelly (1953). They performed acoustoelastic experiments and plotted the stress-dependent shear and bulk moduli, defined as

$$M \equiv \rho v_T^2, \quad K \equiv \rho v_L^2 - \frac{4}{3} \rho v_T^2,$$

against the hydrostatic pressure $P = -\sigma$ for polysterene and for pyrex. For the former, the variations are clearly linear, and nothing would be gained by including FOE effects. For the latter, there is a marked departure from the linear acoustoelastic effect at high pressures, and we thus use (2.8) to obtain a better fit to the data. By digitizing the data and performing a standard least square optimization, we found that for pyrex,

$$M(P) = 2.73 - 8.45 \times 10^{-6} P - 1.21 \times 10^{-9} P^2,$$

$$K(P) = 3.27 - 7.80 \times 10^{-5} P + 4.07 \times 10^{-9} P^2,$$

where $M$ and $K$ are expressed in units of $10^5$ bars and $P$ in bars. The fit to the data is shown in Fig. 1.

According to (2.8), the quantities (2.11) give access to $\lambda$ and $\mu$, to two linear combinations of the third-order constants, and to two linear combinations of the fourth-order constants. Specifically, we may solve (2.11) to find the Lamé constants as

$$\lambda = K(0) - \frac{2}{3} M(0), \quad \mu = M(0).$$
Similarly, we find two linearly independent combinations of the three third-order constants:
\[
A + 3B = -3K(0) [1 + M'(0)] - 4M(0), \quad A + 9B + 9C = -9K(0) [1 + K'(0)] / 2. \tag{2.14}
\]

Finally, we also find two combinations of the fourth-order constants:
\[
E + F + \frac{2}{3}G = \frac{7}{6}K(0) + \frac{4}{3}M(0)
\]
\[
+ \frac{1}{2}K(0) \left[ K'(0) + \frac{13}{3}M'(0) + K'(0)M'(0) + K(0)M''(0) \right],
\]
\[
E + 3F + G + 9H = \frac{17}{24}K(0) + \frac{3}{2}K(0) \left[ K'(0) + \frac{1}{4} (K'(0))^2 + \frac{1}{4} K(0)K''(0) \right]. \tag{2.15}
\]

Hence, for the pyrex data:
\[
\lambda = 1.45 \times 10^5 \text{ bars}, \quad \mu = 2.73 \times 10^5 \text{ bars},
\]
\[
A + 3B = -1.24 \times 10^6 \text{ bars}, \quad A + 9B + 9C = 1.00 \times 10^7 \text{ bars},
\]
\[
E + F + 2G/3 = -1.30 \times 10^7 \text{ bars}, \quad E + 3F + G + 9H = 3.65 \times 10^7 \text{ bars}. \tag{2.16}
\]

Clearly, the data of Hughes and Kelly (1953) are not sufficient to determine all the third- and fourth-order constants. An additional relation is needed to determine experimentally all the third-order constants, and a further two relations to determine the fourth-order constants. These may be provided from, for example, wave speed measurements in uniaxially deformed samples, as we show in the following section.

We conclude this section by evoking the work of P.W. Bridgman on “The Physics of High Pressures” (Bridgman, 1945), which won him the 1946 Nobel Prize. He carried out countless high pressure measurements on solids, and fitted his data with a quadratic equation for the volume change, in the form \( \Delta V/V_0 = -aP + bP^2 \). Direct comparison of this experimental law with (2.3) reveals the identifications \( a = 1/\kappa \) and \( b = -\hat{W}'''(1)/(2\kappa^3) \). Further, acoustoelastic measurements give direct access to these constants, because \( \kappa = K(0) \) and \( \hat{W}'''(1) = -K(0)[2 + K'(0)] \). In Bridgman’s experiments, very high pressures are required to obtain second-order deformations; however, his coefficients \( a \) and \( b \) can also be determined by pressurizing the sample only linearly, and then measuring the speeds of transverse and longitudinal waves. In fact, had Bridgman been able to propagate such waves in his finitely deformed samples he would have had access to the third term in the expansion, \( \Delta V/V_0 = -aP + bP^2 - cP^3 \) say, because the continuation of (2.3) is
\[
\varepsilon = \frac{\Delta V}{V_0} = -\frac{1}{\kappa}P - \frac{1}{2\kappa^3} \hat{W}'''(1)P^2 - \frac{1}{6\kappa^5} \left[ 3 \left( \hat{W}'''(1) \right)^2 - \kappa \hat{W}^{(4)}(1) \right] P^3, \tag{2.17}
\]
where \( \hat{W}^{(4)}(1) \) is found to be expressible as
\[
\hat{W}^{(4)}(1) = \frac{19}{9} \kappa - \frac{4}{9} (A + 9B + 9C) + \frac{8}{3} (E + 3F + G + 9H), \tag{2.18}
\]
in which each term can be determined experimentally using the equations of large acoustoelasticity above.
3 Uniaxial tension

We again consider the cuboidal sample as in Section 2, but now the cuboid is subject to a uniaxial tension in the $X_1$ direction so that it deforms homogeneously with elongation $e_1 = l_1/L_1 - 1 = \lambda_1 - 1$ in the $x_1$-direction. By symmetry it contracts laterally and equibiaxially with elongation $e_3 = e_2 = l_2/L_2 - 1 = \lambda_2 - 1$. The nominal stress (axial force per unit reference area) is $J\sigma_1/\lambda_1 = \sigma_1\lambda_2^2$.

The condition that the lateral faces are free of traction is

$$\sigma_3 \equiv \sigma_2 = J^{-1}\lambda_2 W_2 = 0,$$

which determines the extent of the lateral contraction $-1 < e_2 \leq 0$ (assuming that $\lambda_2 < 1$) and can be written in terms of $e_1$, up to second order, as

$$e_2 = -\alpha e_1 - \beta e_1^2,$$

where

$$\alpha = \frac{\lambda}{2(\lambda + \mu)}; \quad \beta = \frac{3\kappa\lambda}{8(\lambda + \mu)^2} + \frac{\lambda^2 A}{8(\lambda + \mu)^3} + \left[\frac{\lambda(\lambda - 2\mu)}{2(\lambda + \mu)^2} + 1\right] \frac{B}{2(\lambda + \mu)} + \frac{\mu^2 C}{2(\lambda + \mu)^3},$$

and again $\kappa = \lambda + 2\mu/3$ is the infinitesimal bulk modulus. Note that (3.2) collapses to $e_2 = -e_1/2 + 3e_1^2/8$ in the incompressible limits (1.10) and (1.12), as expected from the expansion to second order of the connection $\lambda_2 = \lambda_1^{-1/2}$ between the stretch ratios of an incompressible solid in uniaxial tension.

Substituting (3.2) with (3.3) into the expression $\sigma_1 = J^{-1}\lambda_1 W_1$ for the uniaxial stress, we obtain the relation between the pre-stress and the pre-strain, up to the second order, as

$$\sigma_1 = \frac{3\kappa\mu}{\lambda + \mu} e_1 + \gamma e_1^2,$$

where

$$\gamma = \frac{3\kappa\mu (5\lambda + 3\mu)}{2(\lambda + \mu)^2} + \left[1 - \frac{\lambda^3}{4(\lambda + \mu)^3}\right] A + \frac{3\mu (3\lambda^2 + 4\lambda\mu + 2\mu^2)}{2(\lambda + \mu)^3} B + \frac{\mu^3}{(\lambda + \mu)^3} C.$$

(Note that although $3\kappa\mu/(\lambda + \mu) = E_Y$, the infinitesimal Young’s modulus, we shall not use $E_Y$ hereon.) Conversely,

$$e_1 = \frac{\lambda + \mu}{3\kappa\mu} \sigma_1 - \left(\frac{\lambda + \mu}{3\kappa\mu}\right)^3 \gamma \sigma_1^2.$$

In the incompressible limits (1.10) and (1.12), the stress–strain and strain–stress relations reduce to

$$\sigma_1 = 3\mu e_1 + 3 \left(\mu + \frac{A}{4}\right) e_1^2, \quad e_1 = \frac{1}{3\mu} \sigma_1 - \frac{1}{9\mu^2} \left(\mu + \frac{A}{4}\right) \sigma_1^2.$$

Now we examine the possibility of a small-amplitude body wave travelling in the direction of the unit vector $n$ with polarization in the direction of a unit vector $m$. For a general direction of propagation, the speeds of the different waves are found as the eigenvalues of the acoustical tensor $Q(n)$ in (1.5), i.e. as the roots of a cubic. In this
paper, however, we focus on the wave speeds that are relatively simple to determine experimentally. The speeds of non-principal body waves are not easily accessible because they would require transducers to be placed at an angle to the faces of the cuboid, and not in full flat contact, which would lead to additional transmission problems. This leaves the principal body waves, as explained by Hughes and Kelly (1953): first, waves in the direction $x_1$ of the tension, and, second, principal waves in the $x_2$ and $x_3$ directions, which are equivalent by symmetry.

In the direction of tension there exists a longitudinal wave with speed $v_{11}$, say, and two transverse waves, propagating with the same speed $v_{12}$, say (in fact, these latter two waves may be combined to form a transverse circularly-polarized wave). Hence, $\boldsymbol{n} = \boldsymbol{m} = \boldsymbol{e}_1$ for the longitudinal wave, and $\boldsymbol{n} = \boldsymbol{e}_1, \boldsymbol{m} = \boldsymbol{e}_2$ for the transverse wave, where $\boldsymbol{e}_1$ and $\boldsymbol{e}_2$ are unit basis vectors corresponding to the coordinates $x_1$ and $x_2$, respectively. Using (1.4) and (1.5) we then have

$$\rho v_{11}^2 = J A_{01111}, \quad \rho v_{12}^2 = J A_{01212}. \quad (3.8)$$

Expanding the elastic moduli in terms of the elongation $e_1$, we obtain

$$\rho v_{11}^2 = \lambda + 2\mu + a_{11} e_1 + b_{11} e_1^2, \quad \rho v_{12}^2 = \mu + a_{12} e_1 + b_{12} e_1^2, \quad (3.9)$$

where

$$a_{11} = \frac{\mu}{\lambda + \mu} \left( \lambda + 2B + 2C \right) + 2 \left( 2\lambda + 5\mu + A + 2B \right),$$

$$a_{12} = \frac{\mu}{\lambda + \mu} \left[ 4(\lambda + \mu) + \frac{\lambda + 2\mu}{4\mu} A + B \right], \quad (3.10)$$

for the classical acoustoelastic effect, and

$$b_{11} = \frac{12\lambda^2 + 51\lambda\mu + 34\mu^2}{2(\lambda + \mu)} + \left[ 10 - \frac{\lambda^3}{4(\lambda + \mu)^3} \right] A + \frac{3(\lambda + 2\mu)(12\lambda^2 + 21\lambda\mu + 10\mu^2)}{2(\lambda + \mu)^3} B$$

$$+ \frac{9}{\lambda + \mu} + \frac{\mu^2}{(\lambda + \mu)^3} \right] C - \frac{\lambda^2 A}{2(\lambda + \mu)^3} \left( B + C \right) - \frac{3\lambda^2 + 2\lambda\mu + 2\mu^2}{(\lambda + \mu)^3} BC$$

$$- \frac{3\lambda^2 + 2\lambda\mu + 2\mu^2}{(\lambda + \mu)^3} B^2 - \frac{2\mu^2}{(\lambda + \mu)^3} C^2 + \frac{6(\lambda + 2\mu)}{\lambda + \mu} E + 3 \left( \frac{\lambda + 2\mu}{\lambda + \mu} \right)^2 F$$

$$+ 2 \left[ 6 + \frac{\lambda^2}{(\lambda + \mu)^2} \right] G + \frac{12\mu^2}{(\lambda + \mu)^2} H, \quad (3.11)$$

$$b_{12} = 6\mu + \frac{9(\lambda + 2\mu)}{8(\lambda + \mu)} A + \frac{9\mu}{2(\lambda + \mu)} B - \frac{\lambda^2}{16(\lambda + \mu)^3} A^2 - \frac{3\lambda^2 + 2\lambda\mu + 2\mu^2}{2(\lambda + \mu)^3} B^2$$

$$- \frac{5\lambda^2 + 2\lambda\mu + 2\mu^2}{8(\lambda + \mu)^3} A B - \frac{\mu^2}{4(\lambda + \mu)^3} (A + 4B) C + \frac{3\mu (\lambda + 2\mu)}{4(\lambda + \mu)^2} E + \frac{\mu^2}{(\lambda + \mu)^2} F$$

$$+ \left[ 2 + \frac{\lambda^2}{(\lambda + \mu)^2} \right] G, \quad (3.12)$$
for the large acoustoelastic effect.

For propagation perpendicular to the direction of tension, we take $\mathbf{n} = e_2$. Then there exists a longitudinal wave, with $\mathbf{m} = e_2$ and speed $v_{22}$, a transverse wave polarized in the direction of tension $\mathbf{m} = e_1$ with speed $v_{21}$, and a transverse wave polarized perpendicular to the direction of tension, with $\mathbf{m} = e_3$ and speed $v_{23}$. These speeds are given by

$$\rho v_{22}^2 = J\mathcal{A}_{02222}, \quad \rho v_{21}^2 = J\mathcal{A}_{02121}, \quad \rho v_{23}^2 = J\mathcal{A}_{02323}. \quad (3.13)$$

Expanding the elastic moduli in terms of the elongation $e_1$, we obtain

$$\rho v_{22}^2 = \lambda + 2\mu + a_{22}e_1 + b_{22}e_1^2, \quad \rho v_{2k}^2 = \mu + a_{2k}e_1 + b_{2k}e_1^2, \quad k = 1, 3, \quad (3.14)$$

where

$$a_{22} = -\frac{2\lambda(\lambda + 2\mu) + \lambda A + 2(\lambda - \mu)B - 2\mu C}{\lambda + \mu}, \quad (3.15)$$

$$a_{21} = \frac{(\lambda + 2\mu)(4\mu + A) + 4\mu B}{4(\lambda + \mu)}, \quad (3.16)$$

$$a_{23} = -\frac{\lambda(4\mu + A) - 2\mu B}{2(\lambda + \mu)}, \quad (3.17)$$

are the classical acoustoelastic coefficients, and

$$b_{22} = -\frac{\lambda\mu(\lambda + 2\mu)}{(\lambda + \mu)^2} - \frac{\lambda(2\lambda^2 - \mu^2)}{2(\lambda + \mu)^3}A - \frac{3\mu(3\lambda^2 + 3\lambda\mu + \mu^2)}{(\lambda + \mu)^3}B - \frac{\mu(3\lambda^2 + 4\lambda\mu + 3\mu^2)}{(\lambda + \mu)^3}C$$

$$- \frac{\lambda^2}{4(\lambda + \mu)^3}A^2 - \frac{2(3\lambda^2 + 2\lambda\mu + 2\mu^2)}{(\lambda + \mu)^3}B^2 - \frac{2\mu^2}{(\lambda + \mu)^3}C^2 - \frac{5\lambda^2 + 2\lambda\mu + 2\mu^2}{2(\lambda + \mu)^3}AB$$

$$- \frac{3\lambda^2 + 2\lambda\mu + 6\mu^2}{(\lambda + \mu)^3}BC - \frac{\lambda^2 + 2\mu^2}{2(\lambda + \mu)^3}AC + \frac{3\lambda(\lambda - 2\mu)}{2(\lambda + \mu)^2}E + \frac{3\lambda^2 + 4\mu^2}{(\lambda + \mu)^2}F$$

$$+ 4 \left[ 1 + \frac{\lambda^2}{(\lambda + \mu)^2} \right] G + \frac{12\mu^2}{(\lambda + \mu)^2}H,$$

$$b_{21} = b_{12} - \gamma - \frac{3\kappa\mu^2}{(\lambda + \mu)^2},$$

$$b_{23} = -\frac{\lambda\mu^2}{(\lambda + \mu)^2} + \frac{3(\lambda^2 - \mu^2)}{4(\lambda + \mu)^3}A - \frac{3(3\lambda^2 + 2\lambda\mu + \mu^2)\mu}{2(\lambda + \mu)^3}B - \frac{2\mu^3}{(\lambda + \mu)^3}C - \frac{\lambda^2}{8(\lambda + \mu)^3}A^2$$

$$- \frac{3\lambda^2 + 2\lambda\mu + 2\mu^2}{2(\lambda + \mu)^3}B^2 - \frac{2\lambda^2 + \lambda\mu + \mu^2}{2(\lambda + \mu)^3}AB - \frac{\mu^2}{2(\lambda + \mu)^3}(A + 2B)C - \frac{3\lambda\mu}{2(\lambda + \mu)^2}E$$

$$+ \frac{\mu^2}{(\lambda + \mu)^2}F + \left[ 2 + \frac{\lambda^2}{(\lambda + \mu)^2} \right] G, \quad (3.18)$$

are the ‘large acoustoelasticity’ coefficients.

We may also find expressions for the acoustoelastic effect in terms of the pre-stress $\sigma_1$ as

$$\rho v_{ii}^2 = \lambda + 2\mu + c_{ii}\sigma_1 + d_{ii}\sigma_1^2, \quad \rho v_{ik}^2 = \mu + c_{ik}\sigma_1 + d_{ik}\sigma_1^2, \quad (3.19)$$
where \( i = 1, 2 \) (no sum on repeated \( i \)), and \( k = 1, 3 \) \((k \neq i)\). Using (3.6), we find that

\[
    c_{ij} = \left( \frac{\lambda + \mu}{3\kappa \mu} \right) a_{ij}, \quad d_{ij} = \left( \frac{\lambda + \mu}{3\kappa \mu} \right)^2 \left( b_{ij} - \frac{\lambda + \mu}{3\kappa \mu} \gamma a_{ij} \right),
\]

(3.20)

where \( ij \in \{11, 22, 12, 21, 23\} \). In particular, we recover

\[
    c_{11} = \frac{1}{3\kappa} \left[ \lambda + 2B + 2C + 2\frac{\lambda + \mu}{\mu} \left( 2\lambda + 5\mu + A + 2B \right) \right],
\]

\[
    c_{12} = \frac{1}{3\kappa} \left[ 4(\lambda + \mu) + \frac{\lambda + 2\mu}{4\mu} A + B \right],
\]

\[
    c_{22} = \frac{2}{3\kappa} \left[ B + C - \frac{\lambda}{\mu} \left( \lambda + 2\mu + \frac{A}{2} + B \right) \right],
\]

\[
    c_{21} = \frac{1}{3\kappa} \left[ \lambda + 2\mu + \frac{\lambda + 2\mu}{4\mu} A + B \right],
\]

\[
    c_{23} = -\frac{1}{3\kappa} \left[ 2\lambda + \frac{\lambda}{2\mu} A - B \right],
\]

(3.21)

for the classical acoustoelastic effect (Hughes and Kelly, 1953). Note that \( c_{12} - c_{21} = 1 \) and that \( d_{12} - d_{21} = 1/(3\kappa) \), and hence in the incompressible limit \( d_{12} = d_{21} \). Using these relations, we establish that

\[
    \rho (v_{12}^2 - v_{21}^2) = \sigma_1 + 1/(3\kappa)\sigma_1^2.
\]

(3.22)

Recall now that \( \rho \) is the mass density in the reference configuration. When expressed in terms of the deformed density \( \rho_v = \rho J^{-1} \), equation (3.22) becomes

\[
    \rho_v (v_{12}^2 - v_{21}^2) = \sigma_1.
\]

(3.23)

This relation is in fact exact in accordance with (1.3) and the expressions \( \rho v_{ij}^2 = J A_{\delta_{ij}}, i \neq j \) specialized accordingly with \( \sigma_2 = 0 \).

Finally, we take the incompressible limits of the elastic constants using the limiting values listed in Section 1. There, we have \( \rho v_{11}^2 \to \infty \) and \( \rho v_{22}^2 \to \infty \), unsurprisingly, because longitudinal homogeneous plane waves may not propagate in incompressible solids. For the transverse principal waves travelling in the direction of tension,

\[
    \rho v_{12}^2 = \mu + \left( 3\mu + \frac{A}{4} \right) e_1 + \left( 5\mu + \frac{7}{4} A + 3D \right) e_1^2,
\]

(3.24)

in terms of the elongation (Destrade et al., 2010a), and

\[
    \rho v_{22}^2 = \mu + \left( 1 + \frac{A}{12\mu} \right) \sigma_1 + \frac{1}{9\mu^2} \left[ 2\mu + \frac{A}{4} \left( 3 - \frac{A}{4\mu} \right) + 3D \right] \sigma_1^2,
\]

(3.25)

in terms of the pre-stress. For the transverse principal waves travelling perpendicular to the direction of tension,

\[
    \rho v_{21}^2 = \mu + \frac{A}{4} e_1 + (2\mu + A + 3D) e_1^2,
\]

\[
    \rho v_{23}^2 = \mu - \left( 3\mu + \frac{A}{2} \right) e_1 + \left( 5\mu + \frac{7}{4} A + 3D \right) e_1^2.
\]

(3.26)
in terms of the elongation, see (Destrade et al., 2010a), and, using (3.7),

\[
\rho v_{12}^2 = \mu + \frac{A}{12\mu} \sigma_1 + \frac{1}{9\mu^2} \left[ 2\mu + \frac{A}{4} \left( 3 - \frac{A}{4\mu} \right) + 3D \right] \sigma_1^2,
\]

\[
\rho v_{21}^2 = \mu - \left( 1 + \frac{A}{6\mu} \right) \sigma_1 + \frac{1}{9\mu^2} \left( 8\mu + 3A + \frac{A^2}{8\mu} + 3D \right) \sigma_1^2,
\]

in terms of the pre-stress.

For an application, we use data for a sample of silicone rubber that has been subjected to a standard tensile test. Figure 2 displays the variation of the tensile Cauchy stress component \(\sigma_1\) with the elongation up to a maximum stretch of about 250\%, at which stage one end of the sample snapped out of its grip. Over that range, the TOE strain energy density (1.8) is not able to capture the behaviour of the sample adequately, as shown in Fig. 2, and is thus discarded. On the other hand, the FOE strain energy (1.9) gives an excellent least-squares fit, with coefficient of correlation \(R^2 = 0.9997\). In fact, the fit is very good up to a much larger value of the stretch than could be expected of this fourth-order approximate theory. We determined the following values for the constants of second-, third-, and fourth-order elasticity:

\[
\mu = 109.35 \text{ kPa}, \quad A = -454.18 \text{ kPa}, \quad D = 109.27 \text{ kPa}.
\]

Note that all three are of the same order of magnitude, as expected from the theory (Destrade and Ogden, 2010). We also remark that \(-8\mu < A < -4\mu\), indicating that the corresponding Mooney–Rivlin solid is materially stable (Destrade and Ogden, 2010; Destrade et al., 2010b).

Using these values, the theoretical variations of the squared wave speeds \(\rho v_{12}^2\), \(\rho v_{21}^2\), \(\rho v_{23}^2\) are plotted versus the elongation in Fig. 3. These curves suggest that it is sufficient to elongate the sample by about 20\% to reveal quadratic acoustoelastic effects, and thus to determine \(D\) experimentally.

4 Discussion

This study was set within the framework of the theory of small-amplitude waves propagating in a deformed elastic solid. Existing general expressions for the speeds of body waves in a finitely deformed isotropic elastic solid were recalled and then specialized to fourth-order elasticity. Specifically, the squared wave speeds were expanded in terms of the pre-deformation (and, equivalently, in terms of the pre-stress) in order to reveal the so-called acoustoelastic effect at the considered order. While the classical acoustoelastic effect is concerned with the linear variation of the squared wave speed with the pre-stress, the expansion was extended to the quadratic regime (large acoustoelastic effect). Explicit expressions for the quadratic coefficients for compressible and incompressible materials were determined for the cases of hydrostatic and uniaxial pre-stress. These coefficients give access to the fourth-order elastic constants.

In the case of an incompressible sample under uniaxial tension, for example, measurement of the variations of the speed of a single transverse wave with respect to the pre-stress (or pre-strain) is enough to determine the elastic constants. The wave speed
in the unstressed configuration gives the initial shear modulus $\mu$ of second-order elasticity, the linear variation of the squared wave speed with the pre-stress or pre-strain gives the third-order Landau-coefficient $A$, and the quadratic variation gives the fourth-order constant $D$.

For compressible materials, Hughes and Kelly (1953) showed that any three of the seven equations (2.5), (3.9), (3.14), with just the linear terms retained, or equivalently (2.8), (3.19), excluding either the expression for $\rho v_{12}^2$ or for $\rho v_{21}^2$, suffice to determine the two Lamé coefficients $\lambda$ and $\mu$ of second-order elasticity and the three Landau coefficients of third-order elasticity $A$, $B$, $C$. Here we have shown that any four of these seven equations, with the quadratic terms now included, give access also to the fourth-order constants $E$, $F$, $G$, $H$ (again, excluding either the expression for $\rho v_{12}^2$ or for $\rho v_{21}^2$).

Although we have restricted attention largely to third- and fourth-order elasticity the expressions for the components of the tensor of instantaneous elastic moduli $A_0$ given in (1.2) apply for an arbitrary finite deformation relative to an unstressed configuration of an isotropic elastic material, and the subsequent incremental response depends on the finite deformation and its accompanying pre-stress. If instead there is an initial stress in the reference configuration then the components of $A_0$ are considerably more complicated, as detailed in Shams et al. (2011), but they do clarify how the elastic moduli depend in general on the initial stress. In particular, the dependence of $A_0$ on an initial hydrostatic stress $\tau$ can be put in the simple form

$$A_{0ijkl} = \mu(\tau)(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \lambda(\tau)\delta_{ij}\delta_{kl},$$

where $\mu(\tau)$ and $\lambda(\tau)$ are now stress-dependent Lamé moduli, with $\tau > 0$ ($< 0$) corre-
Figure 3: Plots of $\rho v^2$ (in MPa) versus elongation $e_1$ for a deformed incompressible block of silicone rubber subject to simple tension; theoretical curves for waves travelling (a) in the direction $x_1$ of tension ($v = v_{12}$), (b) in any direction in the plane normal to the direction of uniaxial stress, with transverse polarization in that plane ($v = v_{23}$), (c) in any direction in the plane normal to the direction of uniaxial stress, with polarization normal to that plane ($v = v_{21}$); the dotted lines correspond to the linear part of the $\rho v^2$ versus $e_1$ curves (the classical acoustoelastic effect).

responding to tension (pressure), their precise functional dependence determined by the choice of strain-energy function. We refer to a recent paper by Rajagopal and Sacco-mandi (2009) and references therein for a discussion of the stress dependence of moduli within the context of an implicit theory of elasticity.

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